KYL-20

KYRC

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Date of Issue: July 31, 1953

Report No.: KYL-20

STUDY OF PLUTONIUM AND FISSION PRODUCTS

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STUDY OF PLUTONIUM AND FISSION PRODUCTS

Introduction

A part of the feed material now being processed at the Paducah Plant is uranium that has been recovered from Hanford reactors. This material will be processed in the Feed Plant as well as in the Cascade. Presented in this report are some of the properties of plutonium and the fission products, and a brief review of the available literature regarding their disposition in processes that are conducted at this plant.

As some of the fission products emit considerable penetrating radiation, and plutonium is an energetic alpha emitter, it is felt that this review should be made in order to familiarize the operating and maintenance personnel concerned with some of the conditions which may be encountered.

It is well known that decay products from uranium, UX_1 and UX_2, will be present in major amounts in parts of the Feed Plant and will constitute a major radiation problem. This has not been considered in this report.

Discussion

The Hanford recovered material which will be used at the Paducah Plant will conform to the following specifications as far as plutonium and the fission products are concerned:

All analyses are to be made on a four lot composite from Harshaw and a carload composite from Hanford.

Plutonium - The maximum acceptable concentration is ten parts plutonium per billion parts uranium.

Gamma Activity - The specification for individual composites is that the gamma activity due to fission products is to be less than 300% the gamma activity of aged normal uranium. Likewise, the average of ten successive composites is not to exceed 100% the gamma activity of aged normal uranium.

Beta Activity - The beta activity due to fission products is not to exceed 100% the beta activity of aged normal uranium.

The important fission products which may be contained in the Hanford recovered uranium are as tabulated below:

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Isotope(4)	Half Life	Isotope(4)	Half Life
Ce^{144}	275 d	Sr^{89}	55 d
Cs^{137}	33 y	Te^{127}	90 d
Nb^{95}	35 d	Y^{91}	57 d
Ru^{103}	42 d	Zr^{95}	35 d
Ru^{106}	330 d		

Of these fission products, ruthenium is expected to be present in the largest amount(7). Ruthenium pentafluoride has been prepared from the metal and fluorine at 536° F. (9). Under conditions of operation in the feed plant this compound would be expected to be formed. Ruthenium pentafluoride melts at 223° F. and boils at 595° F. The solubility of ruthenium pentafluoride in uranium hexafluoride is 20.8 mol per cent at 158° F. (1).

From the above properties a large part of the ruthenium would be expected to accompany the uranium through the feed plant and into the cascade. However, due to the extreme reactivity of the pentafluoride with various metals and uranium hexafluoride decomposition products it is more probable that a large part of the ruthenium will deposit in the feed plant equipment. This has been shown to be the case in Oak Ridge pilot and feed plant experiments where a large part of the ruthenium was found in the area between the reactor and the barrier filter (5). Any ruthenium that might be vaporized or mechanically carried over to the cascade from the vaporization baths would be expected to remain near the feed point.

Strontium, cerium, cesium, zirconium, and the rare earths are important fission products that form non-volatile fluorides (7). Therefore, these elements will be found in the ash receiver and barrier filter, or to a lesser extent in any deposits which form in the fluorine towers.

Tellurium hexafluoride is a very volatile compound that would easily be formed in the feed plant. Its boiling point is 32° F. Tellurium tetrafluoride melts at 266° F. and boils at 392° F. Both these compounds react slowly with copper and nickel, forming, in the case of copper, copper telluride and cupric fluoride (8). These reactions are not expected to occur to any extent, and as a result, tellurium should remain with the uranium hexafluoride.

Plutonium hexafluoride has properties very similar to uranium hexafluoride. Figure No. 1 gives vapor pressure - temperature relations. Plutonium oxide is converted to the hexafluoride with fluorine at 1290° F. Plutonium hexafluoride boils at 144.1° F. (2). Uranium hexafluoride sublimes with a pressure of one atmosphere at 133.7° F.

A sample of UAP processed material containing 970 ppb. Pu was fluorinated at different temperatures in an effort to determine the fate of the plutonium during feed plant operations. Approximately 25% of the plutonium was found in the reactor, 5% on the barrier filter, and up to 12% in the uranium hexafluoride under conditions approximating those of the feed plant. The remainder of the plutonium was unaccounted for; however, the data indicates that at lower fluorinating temperatures (750° F.) a higher percentage of the plutonium will be found in the ash. From this experiment and others, it is expected that some plutonium hexafluoride will be formed and will accompany the uranium (5).

Experience in the K-1131 Plant has shown that after running eleven days on Hanford material, plutonium in concentrations up to 70 ppb. can be expected to be found in deposits in the lines leading from the towers. No information was found as to the plutonium content of K-1131 product (10).

In order to determine the fate of plutonium in the cascade, several experiments were performed at K-25. A B-4 pump loop was equipped with a single four foot barrier tube and the loop was charged with uranium hexafluoride made from UAP produced at Oak Ridge National Laboratory. Operating difficulties prevented continuous circulation. Results obtained over a 100 day period failed to show any significant change in the plutonium concentration in the gas phase (6).

An ORNL slug containing 15-20 ppm. plutonium was fluorinated directly from the metal to uranium hexafluoride. The uranium hexafluoride was collected in a cold trap and then transferred directly through a barrier filter into a test loop which consisted of a single barrier tube with a "W" pump for circulation. The cold trap was then washed thoroughly, and an analysis of the uranium in the acid wash solution showed that it contained 10 ppm. of plutonium. Initially the gas samples taken from the loop averaged 0.032 ppm. plutonium. After 50 days the loop was shut down. The barrier contained 0.02 ppm. plutonium. This represents no enrichment over the gas phase plutonium constituent (6).

On the furnace stand at K-25, a size 3 converter with an Allis-Chalmers blower for circulation was charged with uranium hexafluoride made from UAP which was produced at ORNL. This was allowed to circulate 76 days. A statistical analysis of the data showed no significant change in the plutonium concentration. Conclusions based on these experiments were that any plutonium present in the K-25 diffusion plant stream would be in a stable form such that no deposit laid down in any reasonable length of time will ever be distinguishable by direct counting from an equivalent uranium deposit containing no plutonium (6).

Conclusions

Ruthenium may be expected to accompany the uranium during processing in the feed plant except for some concentration of these materials

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in the barrier dust, gas lines from the fluorination towers and the condensation equipment. The ruthenium which accompanies the uranium hexafluoride into the feed cylinders will remain there except for that which is carried into the cascade by entrainment. The amount which enters the cascade will deposit near the feed point.

Any tellurium that is present in the uranium oxide will be expected to accompany the uranium throughout processing.

The fission products strontium, cesium, zirconium, cerium, and the rare earths which form non-volatile fluorides, will concentrate in the reactor trays, ash hoppers of the fluorination towers, and the barrier dust filter. Some entrainment of the non-volatile fission products by the uranium hexafluoride is to be expected. All of these materials which carry over into the cascade should deposit on the barrier surfaces near the feed point. A low degree of concentration of the non-volatile fission products will occur in the feed cylinders and the feed lines to the cascade.

The chemical and physical properties of plutonium indicate that it will accompany the uranium during processing in the feed plant; however, experimental data from small scale fluorinating systems indicate that a low percentage of the plutonium contained in the uranium feed material will accompany the uranium hexafluoride product. This experimental data is inconclusive since much of the plutonium remains unaccounted for. The plutonium which enters the cascade will not deposit to an extent that would create "hot spots".

Recommendations

Although the information available gives a basis for predictions as to the behavior of the fission products and plutonium in the cascade and feed plant, the behavior of these elements should be proven more conclusively by actual sampling and determination of the fission products and plutonium in different places throughout the plant.

The points which should be sampled are the following:

1. The uranium oxide feed material.

2. The barrier filter or cyclone separator between the fluorine tower and cold traps in the feed plant.

3. The uranium hexafluoride as it is emptied from the cold traps to the storage cylinders.

4. If any plutonium or fission products are found in the feed plant product, additional samples should be taken of the cascade feed, tails, and product.

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BIBLIOGRAPHY

1. Farrar, R. L., Jr., Barber, E. F., Capps, R. H., Skidmore, M. R., Bernhardt, H. A., A Review of the Fluorides of Ruthenium, Carbide and Carbon Chemicals Company, K-25 Plant, September 14, 1950 (K-655).

2. Florin, A. E., Plutonium Hexafluoride: Second Report on the Preparations and Properties, Los Alamos Scientific Laboratory of the University of California, November 9, 1950 (LA-1158).

3. Harrison, R. H., McGill, R. M., Powell, E. W., Bernhardt, H. A., Removal of Fission Products from Uranium Tetrafluoride by Fluorination, Carbide and Carbon Chemicals Company, K-25 Plant, July 26, 1950 (K-641).

4. Hume, D. N., Ballou, N. E., Glendenin, L. E., A Manual of the Radiochemical Determination of Fission Product Activities (CN-2815).

5. K-25 Plant Quarterly Report for Second Fiscal Quarter, October 1 - December 31, 1946 (K-560).

6. K-25 Plant Quarterly Report for Third Fiscal Quarter, January 1 - March 31, 1950 (K-600).

7. K-25 Plant Quarterly Report for Fourth Fiscal Quarter, April 1 - June 30, 1950 (K-636).

8. Laboratory Division Progress Report for June 1952, Carbide and Carbon Chemicals Company, K-25 Plant (KLI-625).

9. Mellor, J. W., A Comprehensive Treatise on Inorganic and Theoretical Chemistry, Vol. XV and Vol. XI.

10. Weekly Progress Report Laboratory Division, Carbide and Carbon Chemicals Company, K-25 Plant, August 15, 1952 (KLI-1529-7).

Figure 1